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CAPILLARY CONVECTION WITH SOLIDIFICATION AND
CRYSTAL GROWTH

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INTRODUCTION

In the first phase, experimental and theoretical studies are performed on surface tension-induced natural convection inside sessile drops with internal solidification. Drops of organic liquids and protein (lysozyme) solutions are used. The study is extended to accelerated growth of protein crystal in the second phase. A numerical technique is used to simulate the physical phenomena of solidification process and crystal growth. The goal of this study is to obtain the on-ground information for exploiting the near weightlessness of space to grow protein crystals.

RESEARCH

I. Solidification

Surface tension-induced flow with internal solidification inside a sessile drop is investigated in organic liquids and protein solutions. Both experimental and theoretical studies are performed.

I-a. Experiments

The same experimental apparatus and procedure were employed in both organic liquids [1, 4] and protein (lysozyme) solutions [2, 5, 6]. Figure 1 depicts a schematic of the test setup. It was used to magnify the interior of a sessile drop for viewing the pattern of convective flow (by tracers). After a drop was placed on a glass plate, internal solidification was induced by point cooling at the drop base via a thin copper wire which was connected to a heat sink. The image was picked up by the TV monitor, recorded on video tapes, and displayed in normal or slow motion for detailed studies.

I-b. Theory

A sessile drop is assumed to take the shape of a spherical segment. This postulation is justified in actual observation [7]. Internal solidification inside the drop is induced by point cooling at the center of drop base. Based on the experimental study in Section I-b, the cooling process may be divided into three stages. No phase change occurs in the pre-cooling stage. It is followed by the early stage in which the solid phase grows in a hemispherical form from the drop base center. This stage terminates when the solidification front reaches the drop summit. The final stage of solidification begins thereafter with the solidified phase grows radially outward in a cylindrical form, until the solidification front reaches the drop periphery. The surface tension induced recirculating flow occurs in all three stages. The pattern of recirculating flow varies with the strength of heat removal via the thin copper wire. The speed of solidification front and the flow pattern are theoretically determined. Results are obtained for organic liquids (with transparent solidified phase) and protein solutions. Theory compares satisfactorily with experimental data.

II. Protein Crystal Growth

There is presently a lack of a coherent physical model for protein crystal growth. Another problem is the influence of convection on crystal growth which is still a matter of debate. Some believe convection, especially turbulence, is the major factor in the undesirable incorporation of impurities in crystal growth. Some experiments in space showed that crystals grown in space have better quality and larger dimensions than those grown in terrestrial environment. In a reduced gravity environment, the density-driven

flow is significantly reduced but whether or not the resulting process will be diffusion-limited is still open to debate. The role of Marangoni convection on crystal growth needs to be explored.

II-a. Natural convection effects on crystal growth

An experimental investigation is performed on the growth of protein (lysozyme) crystals in solution layers, a cavity and a vial. The growth process in solution layers is through pure diffusion, while those in the cavity and vial are convection-dominated environments. The convection in the cavity is thermally-induced, whereas that in the vial is solutally-induced. The test setup of Fig. 1 is used. It is concluded that irrespectively of thermally-or solutally-induced mechanism, the effects of convection are [3]

- (i) smaller time constant of growth process,
- (ii) larger average terminal size,
- (iii) shorter terminal time, and
- (iv) lower number flux.

A theoretical model is developed to determine the streamline of recirculation and path of aggregate migration in a cavity. Results agree well with experimental observation.

II-b. Marangoni effects on protein crystal growth

A study is in process to determine Marangoni effects on protein crystal growth. The test apparatus, Fig. 2, has been employed in a preliminary study with success. Currently in process is to grow a protein crystal on a seed surrounded by surface tension-induced recirculation.

PUBLICATIONS

Articles:

1. Y.-J. Su and W.-J. Yang, "Thermocapillary Convection in Evaporating Sessile Drops with Internal Solidification", *Heat Transfer 1990-Jerusalem*, Vol. 4, pp. 233-238, 1990.
2. J.-C. Liu, W.-J. Yang and A. T. Chai, "Micro-Scale Thermocapillary Convection with Solidification", *ASME/JSME Thermal Engineering Proceedings*, Vol 4, pp. 171-177, 1991.
3. J.-S. Lee and W.-J. Yang, "Effects of Natural Convection on Protein (Lysozyme) Crystal Growth in Solution Layer and Cavity", ASME Paper 91-HT-41, 1991.
4. Y.-J. Su, W.-J. Yang and J.-C. Liu, "Mechanics of Transport Phenomena in Multi-Component Sessile Drops with Solidification", *International Communication in Heat and Mass Transfer*, Vol. 17, pp. 729-745, 1990.
5. J.-C. Liu and Wen-Jei Yang, "Thermo-Physical Properties of Lysozyme (Protein) Solutions", accepted for publication in *Journal of Thermophysics and Heat Transfer*,

6. J.-C. Liu and Wen-Jei Yang, "Transport Phenomena in Sessile Drops of Protein (Lysozyme) Solutions with Internal Solidification", to be presented at the Joint Conference of 3rd International Heat Transfer Symposium and 5th International Symposium on Transport Phenomena, October 6-10, 1992, Beijing.

Ph.D. Dissertations:

7. Y.-J. Su, "Thermocapillary Convection in Evaporating Sessile Drops with Internal Solidification, Ph.D. Dissertations, Department of Mechanical Engineering and Applied Mechanics, University of Michigan, Ann Arbor, 1988.
8. J.-C. Liu, "Thermocapillary Convection in Lysozyme Sessile Drops with Internal Solidification, Ph.D. Dissertations, Macromolecular Science and Engineering Program, University of Michigan, Ann Arbor, 1991.
9. K.-S. Huang, "Theoretical Study on Natural Convection in Sessile Drops with Internal Solidification (tentative title)", Ph.D. Dissertation, Department of Mechanical Engineering and Applied Mechanics, University of Michigan, Ann Arbor, 1992 (expected).

Publications No. 1 through 4 are attached.

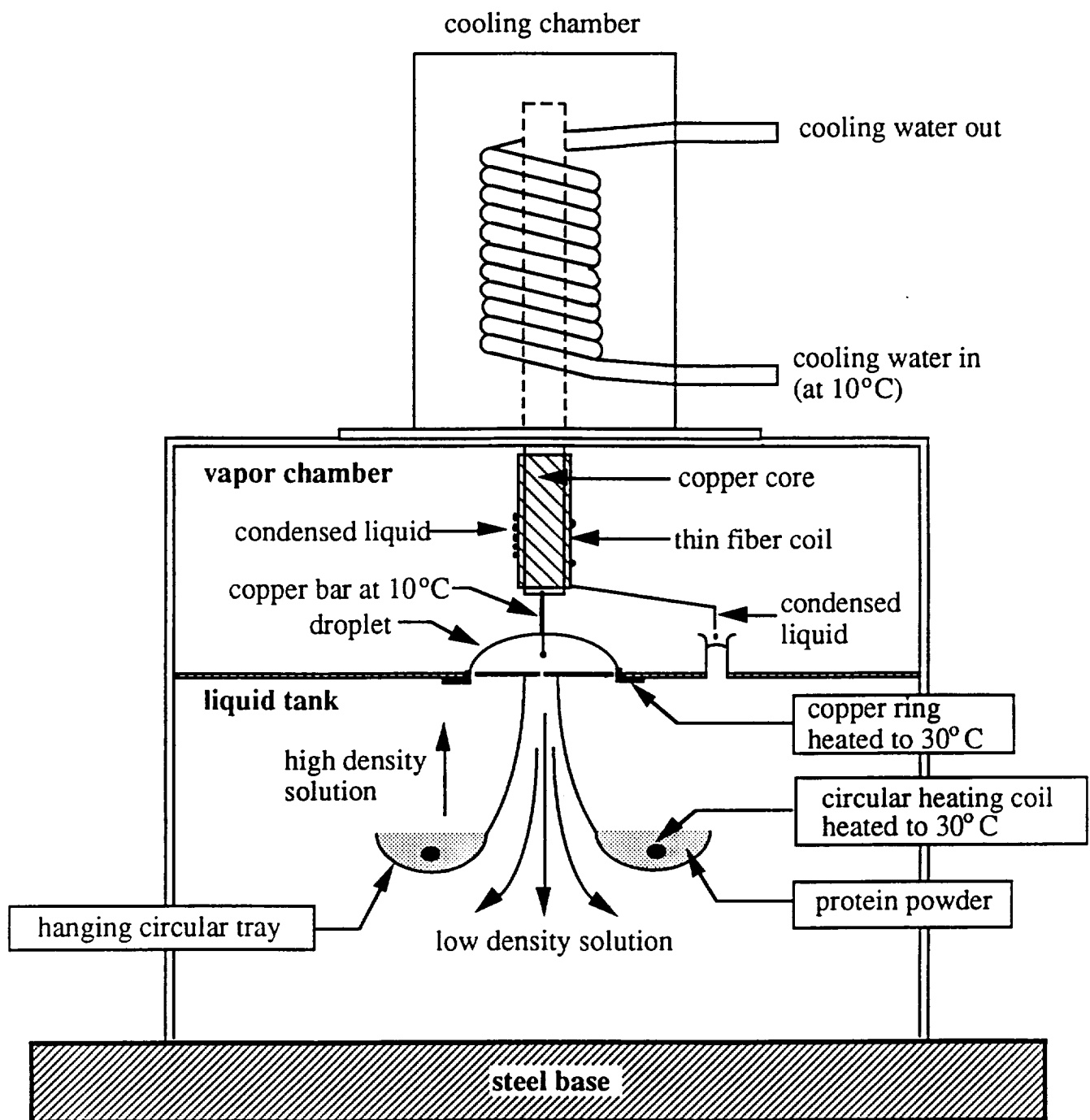


Fig. 2 A schematic diagram of the proposed test setup for growing protein(lysozyme) crystal inside a droplet, simulating reduced gravity environment.